MICROWAVE SENSORS FOR IN SITU EVALUATION OF MOISTURE IN POLYMERS

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INTRODUCTION

We demonstrate the feasibility of a moisture meter which senses the complex permittivity of materials non-destructively. The test materials can be non-conductive solids, particulates or liquids. The meter consists of a sensor plus ancillary electronics. The sensors operate in the microwave region to mitigate certain problems of penetration depth, electrode polarization and ionic conductivity which are encountered in other frequency bands.

One important application of this work is to develop an inexpensive portable probe which can map the moisture contamination *in situ* in the composite skin of an aircraft. Alternatively, the probe can be permanently attached to the inside of the skin to monitor moisture uptake over time. Another interesting application of the same technology is a meter which can monitor and diagnose the cure of the composite.

OPERATING PRINCIPLES

Microwave moisture sensing is based on the large difference in dielectric properties of water relative to most other materials. At 4 GHz, the dielectric constant ε_w' of water is about 76, which is 20 to 30 times higher than the permittivity of most polymers. Similarly the loss factor ε_w'' of free water is quite high, 17 at 4 GHz. As a result, small quantities of absorbed water have a large effect on the microwave dielectric properties of polymers. For frequencies above RF the ionic conductivity does not dominate the loss factor ε'', so the meter reading does not depend on the existence of a concentration of mobile ions. This allows development of a two-parameter system in which both components of the complex permittivity-- the dielectric constant and loss factor --are related independently to moisture content and can be observed simultaneously *in situ*..

In use, the operator presses the sensor against some piece of dielectric material under test (MUT) as in Fig. 1. The meter interrogates the sensor by sweeping the incident microwave energy through a band of frequencies. The meter operates in the reflection mode, in the sense that the spectrum of the wave that is reflected from the sensor contains information about the effective complex permittivity of the MUT[1].

For the applications of interest here, a typical sensor is a cylinder on the order of one inch in diameter and an inch long, with a feed cable and two adjustment screws. See Fig. 2. Electrically, it is essentially an open resonant structure: i.e., an inductively fed dipole or ring microstrip resonator (pat. pending). The microwave fields near the sensor face fringe into the MUT. Any water or wet material in the MUT tends to damp the resonator and reduce its resonant frequency (f_r). The f_r and return loss (L_r), defined by the reflection

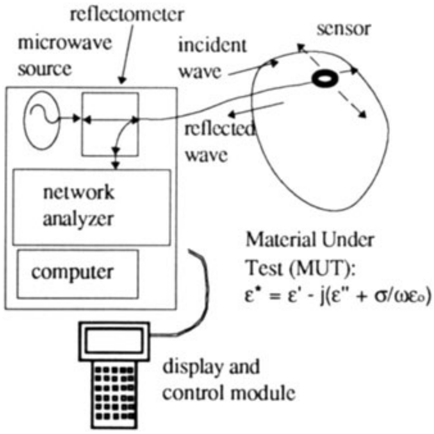

Fig. 1. Block diagram of microwave moisture meter.

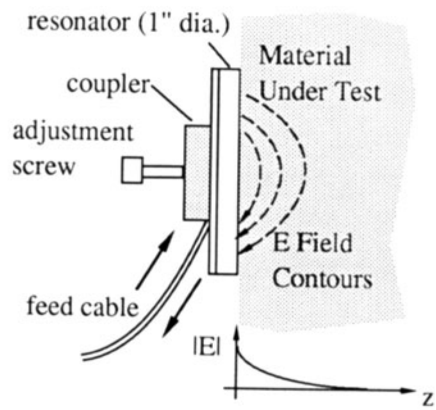

Fig. 2. Schematic drawing of sensor profile.

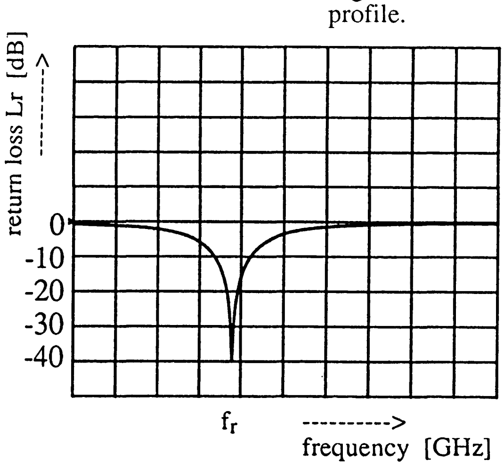

Fig. 3. Typical log magnitude of the sensor's reflected wave vs. frequency

spectrum in Fig. 3, are then used by the meter to calculate the effective ε' and ε" of the MUT. The resonator's size and shape (dipole, ring...) are generally tailored to the application.

DETECTION OF FREE WATER IN HONEYCOMB CELLS

Water can seep into the core of honeycomb via small holes in the skin. One of the potential uses of these sensors is to detect which cell or cells in the honeycomb is contaminated by water. As the sensor is scanned across the skin of the honeycomb, the resonant frequency (f_r) decreases sharply when the sensor is over the contaminated cell.

Epoxy/Glass/phenolic-core Honeycomb

We tested a half-inch thick sandwiched panel consisting of a phenolic honeycomb core with 2-ply epoxy/glass sheeting top and bottom. The core has nine cells to the inch. We used a hypodermic syringe to fill one of the cells with water. A 4.5 GHz dipole sensor was scanned across the wet cell by hand, taking data every 0.1 inch. The dipole was scanned in the direction normal to the dipole orientation, with the wet cell passing under one end of the dipole where the dipole's electric field is the highest.

As seen in Fig. 4, there is a sizable well-resolved spike in f_r when the resonator is directly over the water-filled cell. The sensor does not appear to sense the cell walls, probably because they are very thin. A day later the water had diffused away.

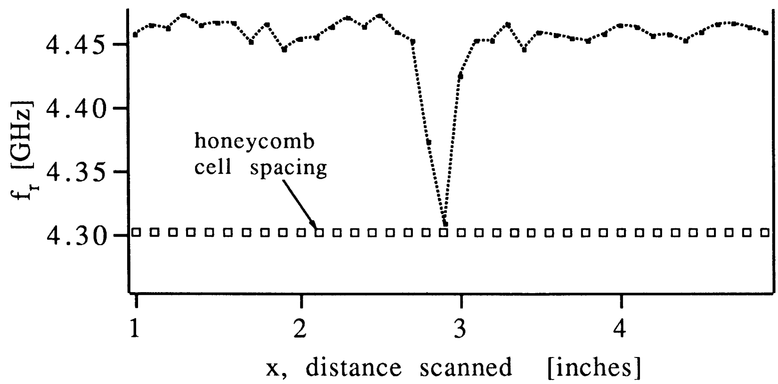

Fig. 4. Detected f_r for free water in cell of epoxy/Kevlar/phenolic honeycomb

Artificial Acrylic Honeycomb

To allow a rapid test of the effect of varying the water level in the cell, we fabricated an artificial honeycomb by drilling an array of holes in a 1/2 inch thick sheet of acrylic. The holes were 1/4 inch in diameter, set on 0.3 inch centers in a rectangular array. We filled one of the holes with dyed water to within about 1/16 inch of the top, then taped a 1/16 inch sheet of acrylic on top of the 'honeycomb' to simulate a thick skin.

A 4.5 GHz dipole sensor was scanned across the acrylic honeycomb as before stopping every 0.1 inch to take data. The results are shown in Fig. 5. In general, the sensor responds most strongly to the objects which are nearest, because the field of the resonator decays exponentially with distance into the MUT. When the cell is completely filled with water, the sensor responds to the bulk of the water in the cell. There is a good-sized spike in resonant frequency when the resonator is directly over the full cell. On the other hand when the cell is only partially filled with water, this particular sensor, having limited penetration depth, responds only to the film of water coating the sides of the cell. Therefore the respective plot of f_r in Fig. 5 has two adjacent dips in f_r. In both cases, the sensor 'sees' the cell walls, as demonstrated by the oscillation in f_r corresponding to the scan over the dry cells.

Oddly, the water is attracted to the sheet covering the honeycomb, perhaps by some electrostatic force. This means that the water 'level' in the cell is not level as depicted in Fig. 5. It is encouraging to see that the dip in the profile of f_r is skewed correspondingly.

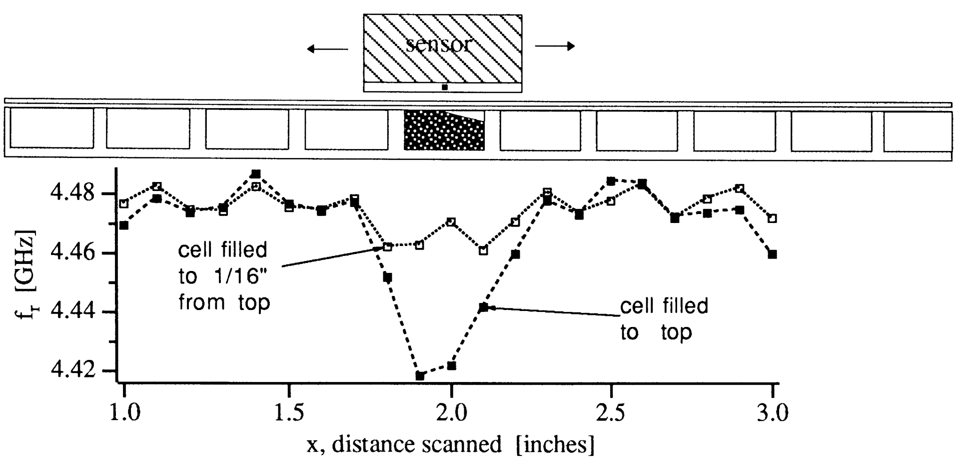

Fig. 5. Detected f_r for free water in cell of acrylic "honeycomb."

To investigate the sensitivity, resolution and linearity of the meter with respect to moisture content (mc), a number of small (approx. 4" x 4") panels or coupons were tested having various resin types and reinforcements, all of which were absorbing moisture at a known rate. The coupons were provided by Hexcel Corp. The baseline moisture content of the coupons is unknown. Several coupons were designated as controls, and sealed in Aclar bags. The rest were immersed in deionized water at 50 $\pm$5°C.

At intervals, each coupon was removed from the hot water, allowed to dry 2 minutes, weighed, tested with a 4.8 GHz. dipole sensor. The test was performed at three different locations on the coupon to average out spatial variations. The sensor was pressed against the coupon under identical conditions (same pressure, same background material, same x-y location, etc.) while recording f_r and L_r. The coupon was then returned to the water. After the coupon had saturated, the changes in weight and electrical data were plotted as a function of the square root of the time. The changes in weight may be regarded, barring erosion of the coupon, as equivalent to changes in moisture content[2].

In Fig. 6 the dielectric constant data (ε') of a neat epoxy resin (Hexcel F161) coupon is compared to the change in moisture content (Δmc). The simulated curve for moisture concentration is derived from Fick's equation with diffusivity D = 5.68 x 10^{-7} cm^2/sec. The abscissa is the square root of the time of exposure to moisture. It is seen that the recorded dielectric constant $\varepsilon'(t)$ tracks mc(t) fairly well. They rise together and tend to level off together. The difference between the two records is attributed to the finite penetration depth of the sensor's electric field. The sensor's response is dominated by the higher concentration near the surface of the coupon. The dashed line overlapping the electrical data in Fig. 6 is the integral of the Fickian moisture content profile, weighted by the exponential electric field profile. Setting the electric field's penetration depth equal to 0.64 mm appears to give the best fit to the ε'' data. Later this estimate was corroborated by a direct measurement of the sensor's electric field. We attribute the small size of the penetration depth to the low dielectric contant of the MUT relative to that of the resonator's encapsulant material. Experimental errors are attributed to the tendency of moisture to out-diffuse from the surface while the measurement was taking place. It is evident that the sensor has the potential to detect unequivocally moisture contents of below 0.1%.

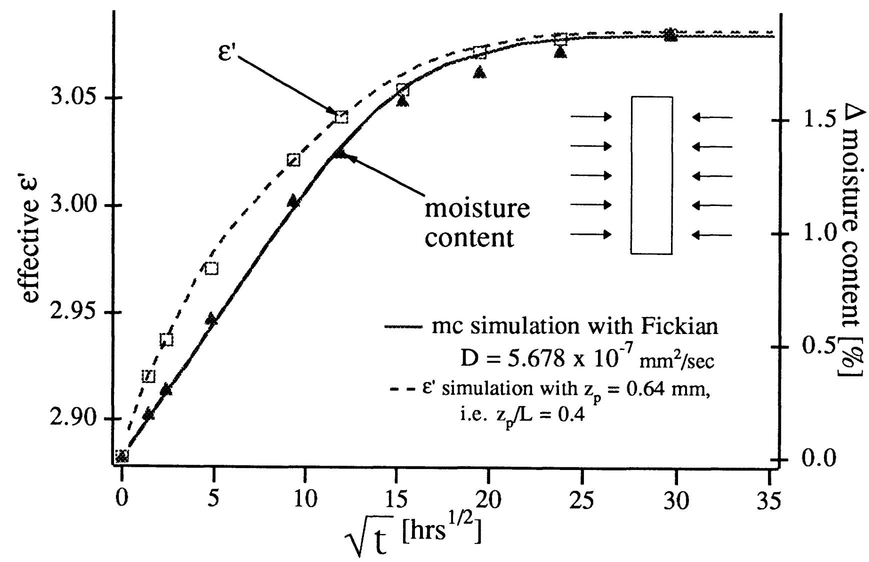

Fig. 6 Moisture content and associated microwave dielectric constant for neat epoxy resin coupon.

LOOKOUT SENSOR

It is not difficult to envision applications of the microwave resonator in which continuous, ongoing measurement of the moisture content would support the intelligent use of some piece of high-value equipment. For instance, in a humid environment, a radome covering a radar antenna would be subject to a gradual change in its dielectric properties. By embedding a sensor in some nonobstructing part of the radome, on the inside looking out, we would be able to monitor the moisture absorption directly in front of the sensor. The moisture absorption of the entire radome could then be related to the electrical signals from the sensor.

Since this "lookout sensor" is fixed in position, we can simplify the construction while enhancing sensitivity and linearity by using the test material as the sensor's substrate. The result is the "smart skin" lookout sensor depicted in Fig. 7. Any moisture which diffuses into the polymer between the resonator and the driving plane enters a region where the electric fields are uniform and very high. The smart-skin version of the lookout sensor is therefore much more sensitive than previous versions of the lookout sensor. Some preliminary data for 1/8 inch epoxy/Kevlar is displayed in Fig. 8. Notice that f_r swings 10 MHz for a change in moisture content of only 0.4%. Also, skin thickness is not a limitation. We have demonstrated that it is possible to inductively couple optimally to a smart-skin sensor when the composite is as thick as 1/2 inch.

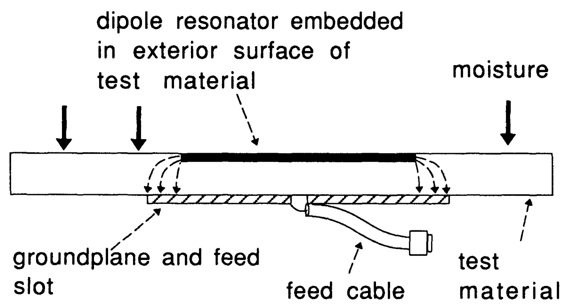

Fig. 7. Sketch of "smart-skin" lookout sensor.

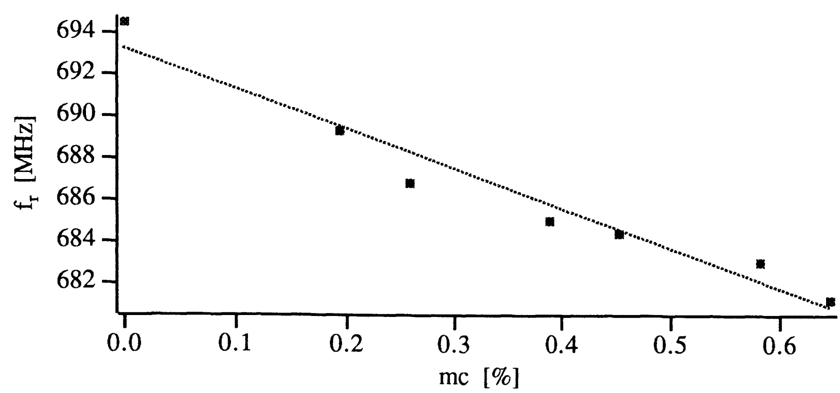

Fig. 8. Data for moisture absorption monitored by "smart-skin" lookout sensor.

Moisture is conducted along the exterior of Kevlar fibers via 'wicking;' that is, some combination of capillary action and diffusion. The fibers have a rough texture similar to cotton. The rate and extent of moisture transport in the direction of the fibers depends on how well the fibers are wetted by the polymer matrix. The effect can be seen vividly by dipping the exposed edge of a Kevlar-reinforced coupon, or for that matter just the edge of the Kevlar cloth by itself, into a pool of dyed water. The color of the coupon or cloth changes behind the diffusion front. Surface or edge wicking can be distinguished qualitatively from bulk wicking by cutting a hole in the coupon before performing the test.

The effect can be quantified using microwave moisture sensors as follows. A rectangular coupon is cut from the test material of interest. Two identical 'smart-skin' lookout sensors are attached to the coupon, as depicted in Fig. 9. The coupon is placed upright with an exposed edge in a shallow pool of water in such a way that conducted water will pass under first one sensor, then the other as the moisture diffuses up the coupon and along the fibers. The sensor responses are monitored as a function of time. The velocity of the diffusion front and the loss of free moisture during diffusion by evaporation or sorption can be determined from a comparison of the sensor responses.

Phenolic/Kevlar&Teflon

Very rapid wicking at room temperature was observed in our first test using Teflon-coated Kevlar in phenolic. The sensors were serpentine dipole resonators positioned about 2 inches apart. In Fig. 10 it is seen that the lower sensor responds first to the diffusing water; the resonant frequency falls sharply. Then, about 8 minutes later, the upper sensor begins to respond. The curves are not linear in $\sqrt{t}$ and are not identical to each other, probably because:

1. there are 2 'hot spots' on each serpentine dipole sensor, i.e. most of the electric field is at the two ends of the dipole and the sensor responds most to the water directly under the ends.
2. the coupon is irregularly shaped instead of rectangular so uniform water flow cannot be expected.
3. the sensors are close enough that they are loosely coupled to each other; i.e. the response of one affects the other's response slightly.
4. water is evaporating as time progresses.
5. the gradient of the diffusion front diminishes as it progresses upward.
6. gravity slows the velocity of diffusion as the height of the capillary 'columns' increases.

As will be seen, similar capillary action occurs in materials of practical interest, so it will be important to isolate and quantify these competing effects.

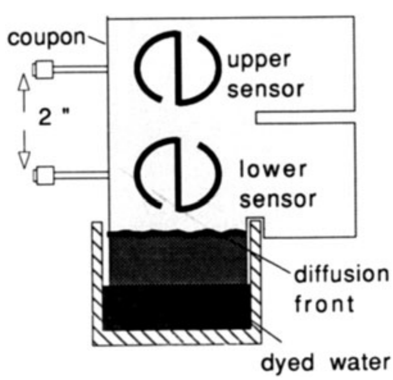

Fig. 9. Sketch of wicking measurement set-up.

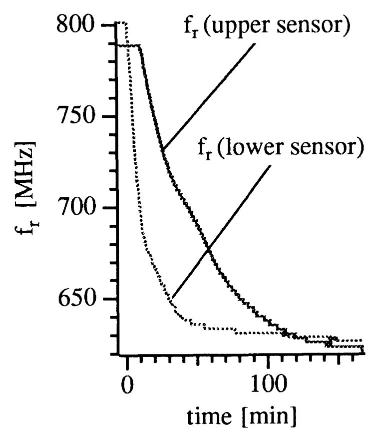

Fig. 10 Data from wicking of moisture in Phenolic/Teflon/Kevlar composite.

Polyester/Kevlar

Somewhat slower wicking was observed in a specimen of Kevlar-reinforced polyester. One side had been painted with some moisture-inhibiting paint. The sensors were ordinary dipoles this time, instead of serpentine dipoles. The dipoles were oriented horizontally in order to reduce the ambiguity in the transit time. They were positioned about 1.6 inches apart on the painted side. The ground plane was glued to the other side, thus forming a version of the "smart-skin" sensor in Figs. 7 and 9.

The diffusion front travelled past the first dipole in about 12 hours, then stopped. The dyed diffusion front was clearly visible between the two dipoles. In Fig. 11 the lower sensor responds to the diffusing water, but the upper sensor does not. Looking at a vertical cross-section of the radome, the water reached its highest level just under the paint on the front surface. It therefore appeared that the diffusion occurred preferentially just under the paint; i.e. the moisture-inhibiting paint appeared to enhance the diffusion rate of the water, well above the diffusion rate on the back surface of the radome. Or, in defense of the paint one could argue that the slower upward diffusion rate on the back side of the radome was due to outward diffusion due to the air (drying). In this view, the paint reduces evaporation from the front surface, allowing the moisture to diffuse to a higher level along the front surface of the radome.

CURE MONITOR PERFORMANCE

These sensors can also be used to monitor the cure *in situ* of an advanced composite panel nondestructively[3]. Fig. 12 compares the cure histories of dry versus moisture-contaminated epoxy/glass prepreg (Hexcel F155/7781). Prepreg is resin that is preimpregnated with fibers. The sensor was flush mounted in a mold such that it pressed against the prepreg. The sensor was a linearly polarized model of the same type used to measure moisture content in Fig. 6.

Both the wet ('mc=0.38%') and 'dry' cures in Fig. 12 were performed at atmospheric pressure in an oven. First the 'dry' cure was performed in which the prepreg had not been exposed to moisture. Then the same sensor was used to monitor the 'wet' cure; another laminate of prepreg from the same bolt had been soaked in 20 °C water for 3 hours, patted dry and exposed to ambient air for 20 minutes before curing. During both cures, the resin temperature was ramped at nearly the same rate, about 4 °C/min, then held at about 121°C. The temperature was monitored by a thermocouple pressed into the prepreg.

As the resin initially heats, its viscosity drops to a minimum, then rises as polymerization and subsequent cross-linking occur. This point of minimum viscosity (PMV) is

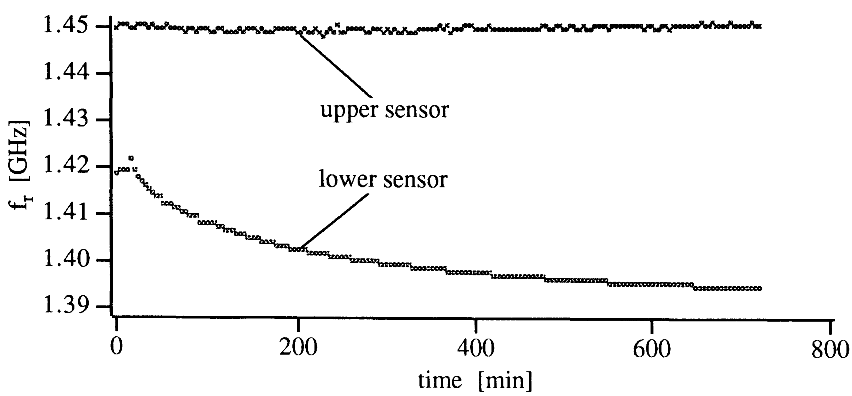

Fig. 11. Data from moisture wicking in polyester/Kevlar composite.

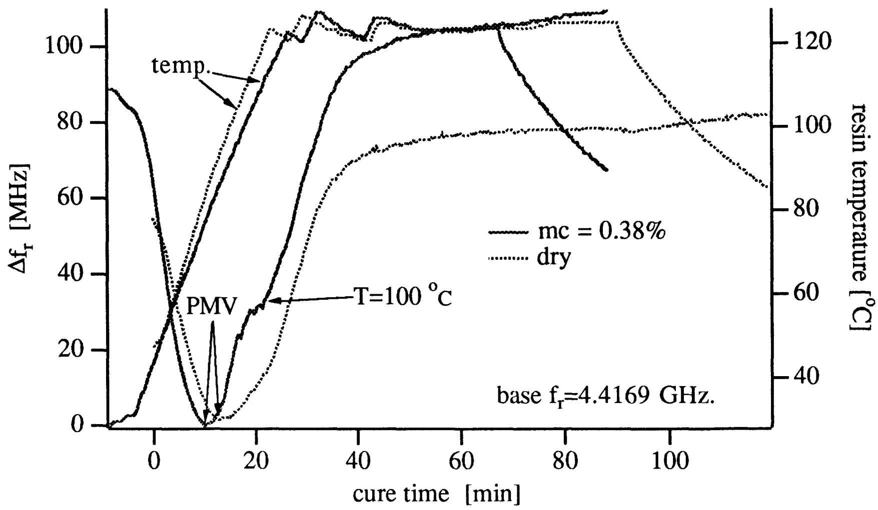

Fig. 12 Cure histories of wet and dry epoxy/glass prepreg at atmospheric pressure.

closely correlated with the minimum of the resonant frequency f_r [2]. Knowledge of the timing of the PMV is of considerable value to a fabricator of composite panels. The PMV for the 'wet' material occurs earlier and at a lower temperature than for the 'dry' prepreg, suggesting that the moisture acts as a catalyst that accelerates the cure process. The upswing in f_r after the PMV provides information about the rate and degree of cure. When the temperature reached 100 °C the moisture in the 'wet' material vaporized, creating erratic variations in f_r. The difference between the asymptotic levels of the 'dry' and 'wet' curves (Δf_r) is associated with a high degree of residual porosity due to water vapor that was observed in the cured 'wet' material.

We could just as easily have monitored the dielectric constant ε', which as a universally recognized property of materials carries more information. But since ε' rises as viscosity falls, a plot of f_r versus time was considered to have more visual appeal to the user.

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